

Giant Rabi splitting in metallic cluster - cavity system

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We investigate theoretically the photo absorption of the cluster of alkali atoms embedded into a single mode quantum microcavity. We show that when the energy of the giant plasmonic resonance lies close to the energy of the cavity mode, the strong coupling between plasmon and cavity photon can occur, which is characterized by mode anticrossing and observation of the doublet structure in photoabsorption. The characteristic values of the Rabi splitting are expected to be several orders of magnitude larger than those observed in single quantum dot- cavity systems.

Introduction Strong coupling between light and matter excitations attract the growing interest of the physical community. The problem is important not only because of the fundamental aspects brought forward by the interaction of material systems with photons [1], but also due to the possibility of use of the strong coupling phenomena for the creation of optoelectronic devices of new generation, such as polariton lasers [2], optical logic gates [3], all-optical integrated circuits [4] and others.

Coupling of zero dimensional (0D) quantum system to a single photon mode, which forms the subject of cavity Quantum Electrodynamics (cQED), is of particular importance from this point of view, because of potential application of cQED to quantum information processing [5–7]. In the domain of the condensed matter, the system which recently attracted particular attention consists of a Quantum Dot (QD) coupled to a single microcavity mode [8–12]. The material excitations in the QD are excitons, that is, bound electron-hole pairs. Owing to their spatial confinement and energy level discretization, they can be brought in strong coupling with the single mode of a microcavity, such as that offered by a pillar (etched planar cavity)[9], the defect of a photonic crystal [10] or the whispering gallery mode of a microdisk [11, 12], among others. In Refs. [9–11], such structures have demonstrated Rabi doublet in their optical spectra, which characterises the mode anticrossing that marks the overcome of dissipation by the coherent exciton-photon interaction. The characteristic values of Rabi splitting for QDs are typically of the order of 10-100 μeV , which is 3-4 orders of magnitude smaller than for the planar microcavities with quantum wells. The small value of the Rabi splitting makes the achievement of the regime of strong coupling in 0D systems technically complicated task and limits the possibilities of its practical implementations.

The natural question arises: can strong coupling regime be observed for material excitations other than excitons? One of the natural candidates is collective plasmonic excitation in metals. For planar metallic structures the dispersion of the surface plasmon lies outside the light cone, which makes its direct optical excitation impossible [13] and rules out the possibility of the observation of any strong coupling effects. This is not true, however, for more complicated structures contain-

ing metallic nanowire arrays [14] and metallic nanorods [15], for which the effects of strong coupling were shown to be extremely pronounced and experimentally observed Rabi splitting can be as big as 250 μeV [14]. This number exceeds by the order of magnitude the values of Rabi splitting for exciton-photon coupling in planar inorganic microcavities [16] and is comparable to Rabi splittings observed in organic structures [17].

In the present work we consider another type of hybrid metal-dielectric structure, consisting of the single metallic cluster embedded inside a single mode photonic cavity. The collective motion of electrons in the cluster against the positively charged ionic background leads to formation of a surface plasmonic mode responsible for the appearance of a giant resonance in the photoabsorption spectra [18–22]. If the energy of the plasmon is close to that of the photonic mode of the cavity, the processes of multiple resonant emissions and absorptions of photons by the cluster can take place and hybrid plasmon-photon modes be formed. The goal of the present letter is to analyze their influence on photoabsorption spectrum of the system and to estimate the corresponding values of the Rabi splitting.

The model. To illustrate the onset of the strong coupling regime in cluster- cavity system we will focus on the metallic clusters formed by alkali atoms (Li, Na, K), as they are the most exhaustively studied from both theoretical and experimental points of view. The geometry of the system we consider is shown on Fig.1

The theoretical basis of studying of the collective phenomena in clusters is provided by the jellium model, in which N electrons (one per each alkali atom) are moving in the spherically symmetric electrostatic potential $V_b(r)$ formed by the uniform background of the positive ions, calculated as convolution of the Coulomb electrostatic potential with smooth charge distribution,

$$V_b(r) = \begin{cases} -\frac{Ne^2}{8\pi\epsilon_0 R} \left[3 - \left(\frac{r}{R} \right)^2 \right], & r \leq R, \\ -\frac{Ne^2}{4\pi\epsilon_0 r}, & r > R \end{cases} \quad (1)$$

This assumption is generally believed to be well justified for the clusters with closed shells [23], i.e. for $N = 8, 18, 20, 34, 40, 58, 92, \dots$, and in the present work we will assume that this condition is satisfied. We underline, however, that our results are of general char-

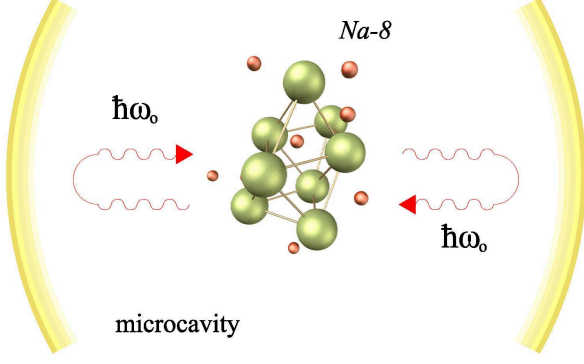


FIG. 1: Metallic cluster formed by alkali atoms Na-8 in microcavity. The cavity photons of the frequency ω_0 undergo multiple reemissions and reabsorptions by a collective excitation of the cluster forming light-matter hybrid polariton eigenmodes of the system.

acter and should be qualitatively the same for clusters with partially filled shells and for clusters consisting of the non-alkali atoms. The computation procedure will become more tricky in these latter cases [24].

Let us consider an alkali-metal cluster with transition frequencies $\omega_{\alpha j} = \omega_j - \omega_\alpha = \hbar^{-1}(E_j - E_\alpha)$ where the indices α and j correspond to the occupied and unoccupied single electron states respectively. In the case when electron-electron interactions are neglected or treated within Hartree-Fock approximation, each single electron transition is coupled to the external photons independently. As oscillator strengths of these individual transitions are small, their coupling with cavity mode will result in low values of Rabi splitting (of the order of magnitude of those observed in QD-cavity systems).

However, the account of the many-body corrections can change the picture dramatically. Coulomb interaction can lead to the formation of the collective excitation of the cluster known as giant plasmonic resonance and having huge oscillator strength (several orders of magnitude bigger than oscillator strengths of individual transitions) [18–22]. Consequently, one can expect high values of the Rabi splitting in coupled cluster-cavity system.

Formalism. The interaction of the electromagnetic field with material objects is described by dipole matrix elements of the transitions $d_{\alpha j}$. The account of many electron processes results in renormalization of dipole matrix elements which become frequency-dependent and are denoted as $D_{\alpha j}(\omega)$ in our further discussion.

In diagrammatic representation, the equation for $D_{\alpha j}(\omega)$ for the system we consider is shown at Fig.2, and graphical representations of renormalized and bare dipole matrix elements of photoabsorption $D_{\alpha j}(\omega)$ and $d_{\alpha j}$ on Fig.3. At the right hand side of Fig.2 the first diagram corresponds to the direct photoabsorption, the second two diagrams accounts for the influence of dynamical polarizability in random phase approximation with exchange (RPAE) and give rise to giant plasmon resonance, and the last one accounts for the multiple re-emissions and re-absorptions of the cavity photon. The

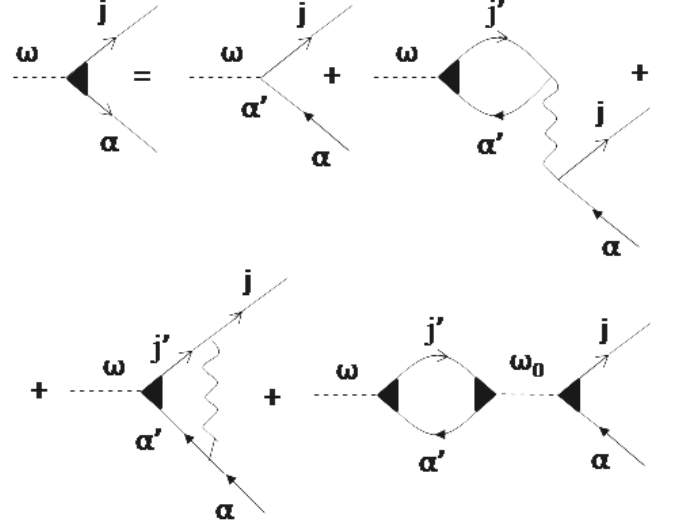


FIG. 2: Diagrammatic representation of the equation for the renormalized dipole matrix element of the transition. The straight lines with arrows correspond to the electrons in the cluster at vacant and filled orbitals j, α , the wave lines to the Coulomb interaction, the dotted lines - to the cavity photons of frequency ω_0 .

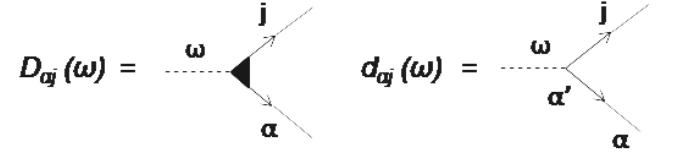


FIG. 3: Diagrammatic representation of renormalized and bare dipole matrix elements of the transition

straight lines with arrows correspond to the electrons in the cluster at vacant and filled orbitals j, α , the wave lines to the Coulomb interaction, the dotted lines - to the cavity photons of frequency ω_0 .

Using the standard rules of the evaluation of the Feynmann diagrams, one can easily find that renormalized dipole matrix elements can be found from the following set of non-linear algebraic equations:

$$D_{\alpha j}(\omega) = d_{\alpha j}(\omega) + \sum_{\alpha', j'} M_{\alpha', \alpha'; j, j'}(D(\omega)) D_{\alpha' j'}(\omega) \quad (2)$$

where

$$M_{\alpha', \alpha'; j, j'}(D(\omega)) = \frac{\langle \alpha', j' | V_C | \alpha, j \rangle - \langle \alpha, j' | V_C | \alpha', j \rangle}{\omega - \omega_{\alpha' j'} + i\Gamma} + (3) \\ + \frac{2\omega_0 g_{\alpha' j'}^* (D_{\alpha' j'}(\omega)) g_{\alpha j} (D_{\alpha j}(\omega))}{(\omega^2 - \omega_0^2 + 2i\omega_0 \gamma) (\omega - \omega_{\alpha' j'} + i\Gamma)}.$$

In the above formula $\langle \alpha', j' | V_C | \alpha, j \rangle$ denotes the matrix element of the Coulomb interaction between the electrons, the linewidths Γ and γ correspond to the finite lifetime of the one-electron excitations and cavity photons, $g_{\alpha j}$ are the matrix elements of the interaction between the transition $\alpha \rightarrow j$ and cavity mode, which are proportional to the renormalized dipole matrix elements

of the transitions and can be estimated as [25]

$$g_{\alpha j} \approx \omega_{\alpha j} \sqrt{\frac{\hbar}{2\varepsilon_0\omega_0 V}} D_{\alpha j}(\omega) \approx \omega_{\alpha j} \omega_0 \sqrt{\frac{\hbar}{2\pi^3\varepsilon_0 c^3}} D_{\alpha j}(\omega) \quad (4)$$

where $V \approx (\lambda_0/2)^3$ denotes the cavity volume.

Note, that solving the system of equations (2) is equivalent to accounting the Coulomb RPAE diagrams and coupling between transitions in cluster and cavity mode up to the infinite order. Together, they describe the formation of the hybrid plasmon- photon excitations in cluster- cavity system.

The response of the system (or photoabsorption coefficient) is proportional to the imaginary part of the dipole dynamical polarizability

$$\sigma(\omega) = 4\pi \frac{\omega}{c} \text{Im} [\alpha(\omega)] \quad (5)$$

which can be found using standart formula

$$\alpha(\omega) = -e^2 \sum_{\alpha j} \left[\frac{|D_{\alpha j}(\omega)|^2}{\hbar\omega - \hbar\omega_{\alpha j} + i\Gamma} - \frac{|D_{\alpha j}(\omega)|^2}{\hbar\omega + \hbar\omega_{\alpha j} + i\Gamma} \right] \quad (6)$$

The energies of the eigenstates of the system can be found from the poles of the dressed Green function for the cavity photon, represented diagrammatically on Fig. 4. The double dashed line corresponds to the renormalized Green function of the photon, the single dot line- to the Green photon of the bare photon $G^0 = 2\omega_0/(\omega^2 - \omega_0^2 + 2i\omega_0\gamma)$. The dot on the diagram corresponds to the renormalized matrix element of the cluster- cavity coupling $g_{\alpha j}(D_{\alpha j}^{(0)}(\omega))$, accounting for many- body interactions in the cluster but neglecting multiple re-emissions and re-absorbtions of the cavity photon, as latter processes are already accounted for in the diagram for in the Dyson equation for the dressed photon (Fig.4) and should not be counted twice. Mathematically, the values of $D_{\alpha j}^{(0)}(\omega)$ can be found from the system of the equations analogical to those represented in diagrammatic form at Fig.2, but with last diagram in the right hand side being absent. The Green function of the dressed photon reads

$$G_{ph} = \frac{1}{(G_{ph}^0)^{-1} - \Sigma(\omega)} \quad (7)$$

where

$$\Sigma(\omega) = \sum_{\alpha j} \frac{|g_{\alpha j}(D_{\alpha j}^{(0)}(\omega))|^2}{\omega - \omega_{\alpha j} + i\Gamma} \quad (8)$$

The eigenfrequencies of the hybrid eigenmodes can be found from the poles of the expression 7 giving the following transcendent equation:

$$\omega^2 - \omega_0^2 + 2i\omega_0\gamma - 2\omega_0\Sigma(\omega) = 0 \quad (9)$$

Before we proceed with presenting the results of the numerical modelling of our system, let us analyze some simple limiting cases.

First, if the cavity is absent, electron- electron interactions renormalize dipole matrix elements of the transitions $D_{\alpha j}^{(0)}(\omega)$ which become frequency dependent and

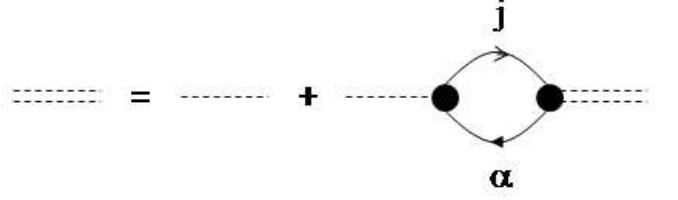


FIG. 4: Diagrammatic representation of Dyson equation for the Green's function of the cavity photon. The double dashed line corresponds to the renormalized Green function of the photon, the single dot line- to the Green photon of the bare photon. The dot on the diagram corresponds to the renormalized matrix element of the cluster- cavity coupling accounting for many- body interactions in the cluster but neglecting multiple re-emissions and re-absorbtions.

form sharp peaks at the frequency $\omega = \omega_{pl}$ corresponding to the characteristic frequency of giant plasmon resonance [18–22] (see inset on Fig. 5).

On the other hand, if one neglects Coulomb interactions (which corresponds to the retaining of the first and last diagrams only in the diagrammatic equation presented on Fig.2) and considers the coupling of the individual single transition to the cavity mode, the equations for renormalized dipole matrix element and dressed photonic Green function can be easily solved and read:

$$D_{\alpha j}(\omega) = \frac{d_{\alpha j} (\omega^2 - \omega_0^2 + 2i\omega_0\gamma) (\omega - \omega_{\alpha j} + i\Gamma)}{(\omega^2 - \omega_0^2 + 2i\omega_0\gamma) (\omega - \omega_{\alpha j} + i\Gamma) - 2g^2\omega_0} \quad (10)$$

$$G_{ph} = \frac{2\omega_0 (\omega - \omega_{\alpha j} + i\Gamma)}{(\omega^2 - \omega_0^2 + 2i\omega_0\gamma) (\omega - \omega_{\alpha j} + i\Gamma) - 2g^2\omega_0} \quad (11)$$

The poles of these expressions determine the energies of the new hybrid eigenstates of the system. The condition of achievement of the strong coupling regime characterized by the mode and anticrossing at the resonance ($\omega_0 = \omega_{\alpha j}$) is given by a standard expression $4g^2 > (\Gamma - \gamma)^2$ [26].

Results. Now let us present the results of the numerical modelling of the realistic cluster- cavity system. We consider a Na-8 cluster embedded in a photonic cavity in the position where the electric field of the cavity mode reaches its maximum. The energy of the cavity mode ω_0 is supposed to be close to the energy of the giant plasmon resonance ω_{pl} . The energies of the single electron states in the cluster were calculated within jellium model using Hartree- Fock approximation. The non-radiative linewidths of all individual transitions in the cluster were taken to be the same and correspond to $\Gamma = 0.8$ meV. The nonlinear system of equations (2) and 9 for the dipole matrix elements and eigenfrequencies of the system were solved using iterative procedure of successive approximations.

Fig.5 shows the absorbtion of the cavity cluster system as a function of frequency of the external laser excitation for the case when the energy of the giant plasmon is tuned in resonance with the energy of the photonic mode, $\omega_0 = \omega_{pl}$. If the lifetime of the photons in microcavity τ is low ($\gamma \approx \hbar/\tau$ is high) we can observe one peak on the photoabsorption spectrum (see Fig. 5, note, all the plots are normalized to unity). However, the situation qualitatively changes if lifetime increases. In this case the strong coupling regime is established and two peaks

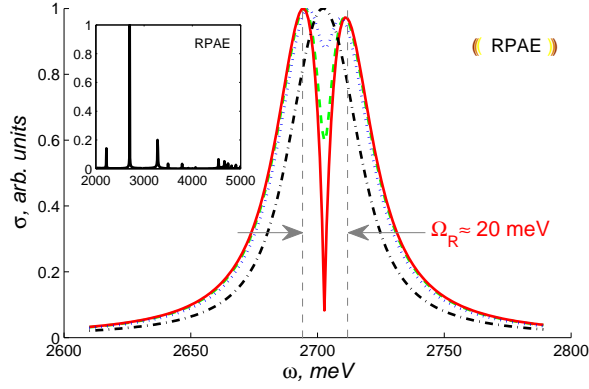


FIG. 5: Photoabsorption cross-section spectrum - system response on the exciting emission of light for different lifetimes of the photons (in meV): 0.1 (red/solid), 2 (green/dashed), 4 (blue/dotted) and 8 (black/dash-dotted). Rabi splitting Ω_R decreases at the photons lifetime being decreased and finally disappears at $\gamma \approx 8$ meV. Inset shows spectral characteristic of the renormalized dipole matrix element of the cluster Na-8 (without a cavity). See text for the details.

are observed in photoabsorption. The distance between the peaks corresponding to the Rabi splitting increases with increase of the lifetime of the cavity mode and can reach the values of about 20 meV. This is several orders of magnitude bigger than the numbers observed in single QD- cavity systems [8–12].

Fig. 6 shows the dependence of the real parts of the eigen frequencies on the cavity energy ω_0 for different linewidths of the cavity (in meV). The Rabi splitting can be found as distance between the two branches at the anticrossing point corresponding to $\omega = \omega_0$ and $\omega = \omega_{pl}$ and it varies from ≈ 0 meV ($\gamma = 8$ meV) to 17 meV (for $\gamma = 0.1$ meV). The inset illustrates dependence of splitting on γ . Note that the results presented on Fig. 5 and 6 are in good agreement with each other. It should be noted that the increase of the number of the atoms in the cluster N leads to the increasing of the Rabi splitting. This increase, however is non proportional to N - our estimations give maximal values of about 25 meV for Na_{20} cluster (as compare to about 20 meV for Na_8 cluster).

Conclusions. In conclusion, we have analyzed the spectrum of photoabsorption of an individual Na-8 cluster embedded in single mode photonic microcavity. We have shown that in the region of the giant plasmonic resonance the regime of the strong coupling between plasmon and cavity photon can be achieved, which manifests itself by formation of the Rabi doublet and mode anticrossing. A cluster can be considered as a 0D object with size comparable to those of the QDs. However, due to the many-body effects it demonstrates Rabi splittings several orders of magnitude bigger than other 0D quantum objects.

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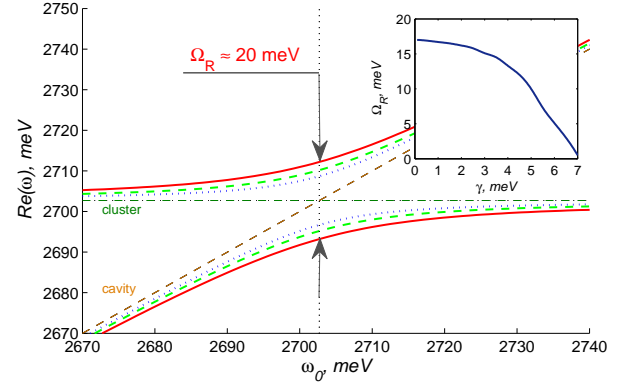


FIG. 6: Eigen frequency real part dependence on the microcavity mode wavelength for different lifetimes of the photons (in meV): 0.1 (red/solid), 2 (green/dashed) and 4 (blue/dotted). Illustration of the crossover from strong- to weak-coupling regime. When $\gamma \approx 8$ meV the splitting $\Omega_R = 0$ and one can see two solutions corresponding to the bare photon and plasmon $\omega_1 = \omega_0$ and $\omega_2 = \omega_{cl}$.

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